



## Review article

# Integrity of aquatic ecosystems: An overview of a message from the South Pole on the level of persistent organic pollutants (POPs)<sup>☆</sup>



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## ARTICLE INFO

**Keywords:**

Aquatic ecosystems

Level of integrity

Persistent organic pollutants (POPs)

## ABSTRACT

Two third of our planet is covered by water, and lakes and rivers are the most important freshwater resources. But the amount of fresh water represents only 2.5% of the earth's water. Of the earth's fresh water, about 70% is locked up in the continental ice, 30% in underground aquifers and only 0.3% in rivers and lakes. In many water ecosystems around the world, climate change and environmental contamination affect water quantity and quality and water-related ecosystems. It is well known that a fifth of the population in Europe and the America drink contaminated water which does not meet international standard criteria, and also that the consumption of low-quality drinking water is responsible for around 80% of human diseases in the world (official data from World Health Organization, WHO). Thus, the assessment of the integrity of aquatic ecosystems is crucial for its correct management. One of the most critical parameter in evaluating the water resource integrity is the contamination level that can be evaluated by comparing it with equivalent ecosystems in their "natural" (or "pristine") state. However, because of the global impact of human activities, only Antarctica can be still considered "almost natural".

Persistent Organic Pollutants (POPs) represent one of the most studied class of environmental contaminants due to their well-known carcinogenic and mutagenic effects on living organisms. They are ubiquitous, lipophilic and accumulate in lipid tissues and trough food chains. In this work an overview on the presence of POPs in several aquatic ecosystem around the world will be comparatively discussed with data obtained at the South Pole of our planet. Moreover, the present trend of the presence of POPs at a global level will be evaluated on the basis of concentration profiles as obtained from ice-cores gathered at Victoria Land, Antarctica. Finally, some conclusive remarks are presented on the use of the information available in the management of water resources.

## 1. Introduction

The integrity of an aquatic ecosystem should be evaluated on the basis of a well-established set of chemical, physical and biological characteristics [1]. Ecological integrity was firstly mentioned in the Clean Water Act (CWA) released by the U.S. Congress on 1972 [2–4], that gave to the U.S. Environmental Protection Agency (US-EPA) the authority to implement pollution control programmes. In Europe, the EU Water Framework Directive (WFD) was released on 2000 and addressed for the first time in an integrated manner the water management. It made clear that water resource integrity included land-use and management, besides all aspects dealing with water distribution and treatment [5]. According to the World Health Organization (WHO) [6] a fifth of the population in Europe and the America drink contaminated

water which does not meet international standard criteria, and also that the low integrity of water resources for human consumption is responsible for around 80% of human diseases in the world [7].

From an operational point of view, in order to evaluate the evolution of an ecosystem we refer to the level of integrity that is determined by all available information at a predefined point on the time scale taken as reference. In this regard, the path to a conscious management of water resources should be based on an integrated aquatic ecosystem management, and should ensure proper planning and implementation of two very important tasks: coordinated monitoring activities to characterize the aquatic ecosystem, and a comprehensive data analysis to evidence trends of the level of ecological integrity [5,8,9] (Fig. 1).

If a negative trend is evidenced, two more difficult tasks should be addressed: source identification and apportionment, to possibly

\* Selected papers from the XVI Hungarian – Italian Symposium on Spectrochemistry: Technological innovation for water science and sustainable aquatic biodiversity, 3–6th October 2018, Budapest.

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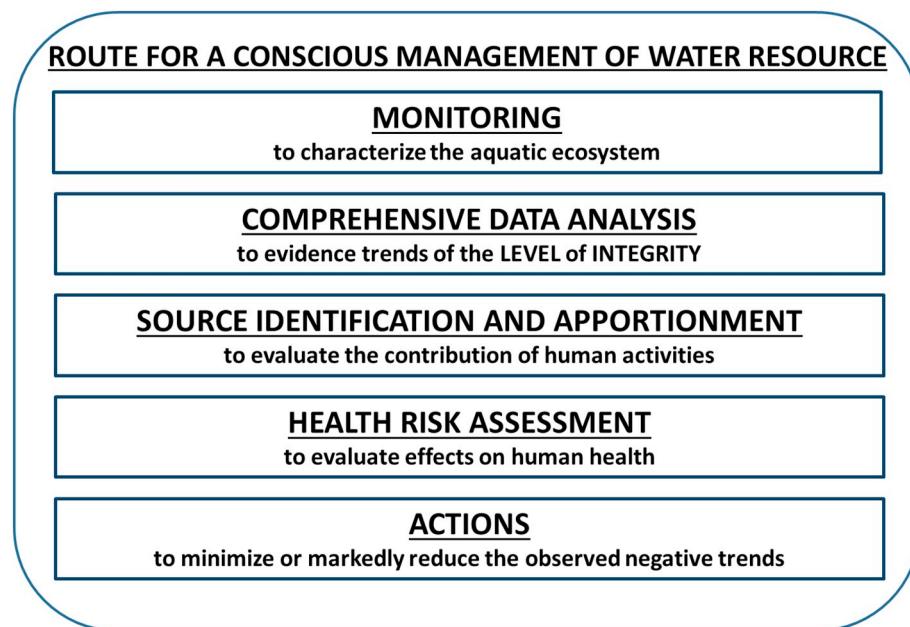


Fig. 1. A schematic description of the route for a conscious management of water resources.

evaluate the contribution of human activities, and health risk assessment, to evaluate the effects on human health. In case a significant risk for human health is evidenced, we finally come to the most difficult step: actions to minimize or markedly reduce the observed negative impacts. The last step is largely entrusted to the politic and economic bodies at various levels (local, regional, national and international) [10–13].

Climate change, eutrophication and chemical contamination, as a result of human activities, have to be included in the assessment of the integrity of a water ecosystem in order to guarantee a more environmentally sensitive and ecologically sustainable management of the water resource [8]. As far as chemical pollution is concerned, dissolved oxygen, nutrients, minerals and contaminants are the most important chemical parameters for water quality assessment. A water ecosystem can be contaminated by direct and indirect sources [14]. Direct sources are mainly associated with industrial and agricultural activities whose waste effluents directly contaminate the sources of water supply destined for human consumption. Indirect sources are associated with the transport and diffusion processes of contaminants from soil and atmosphere to water ecosystems [15,16].

This paper takes into consideration the chemical contamination of water resources, particularly the class of persistent organic pollutants (POPs), as it is included among the most important parameters in assessing the integrity of the water ecosystem. A concise review on the world-wide presence of POPs, the effects on human health, the source identification and apportionment, and the message from the Antarctica on the temporal trends of these contaminants are presented. Finally, some conclusive remarks are outlined on the use of the information available that policy makers should make to plan incisive actions for limiting environmental contamination. These actions should be the basis of a responsible management of water resources and are mandatory to ensure an adequate level of integrity.

## 2. The chemical contamination of aquatic ecosystems

The environmental concern on chemical pollutants is mainly related to the inadequacy of: (i) the available tools for assessing the effects both on the biodiversity in an aquatic ecosystem and on the human health, (ii) the technologies for remediation and water-treatment, (iii) the strategies for usage and disposal [16]. Among organic contaminants,

persistent organic pollutants (POPs) include many classes of organic compounds that are of great importance in the assessment of environmental contamination due to their toxic effects on human health, often carcinogenic and/or teratogenic in nature. Moreover, POPs are generally very stable and are resistant to environmental degradation through biological, chemical and photolytic processes [17,18]. After entering the environment, they can travel long distances in the atmosphere before deposition occurs [19–22]. As a consequence, these compounds undergo global fractionation, and finally may accumulate to the poles by cold-trapping process [22–24].

Once the effects on human health have been assessed, various international initiatives have been launched since the 1990s aimed at reducing and/or eliminating emissions and discharges of POPs, recognizing the need for global actions to better protect and safeguard human health and the environment.

At present, POPs have been considered in several internationally accepted protocols and conventions. Among them, the following two can be cited:

- the Stockholm Convention on POPs [25] signed on 2001 and entered into force on 2004. The Convention includes the following 12 pollutants or class of pollutants: aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins and dibenzofurans. On 2009 the Conference of the Parties (COPs) considered a recommendation from the POP-Review Committee to add the following nine chemicals in the Annexes of the Convention: chlordcone, hexabromobiphenyl, tetra-, penta-, hexa- and heptabromodiphenyl ether,  $\alpha$ -hexachlorocyclohexane,  $\beta$ -hexachlorocyclohexane,  $\gamma$ -hexachlorocyclohexane (lindane), pentachlorobenzene, perfluorooctane sulfonic acid (its salts and perfluorooctane sulfonyl fluoride);
- the United Nations Economic Commission for Europe (UNECE) protocol on POPs [26] which was signed on 1998 and entered into force on 2003. The protocol includes the following 16 POPs: aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins, dibenzofurans, chlordcone, hexachlorocyclohexane (including lindane  $\gamma$ -HCH), hexabromobiphenyl and polycyclic aromatic hydrocarbons (PAHs).

POPs can be roughly classified in two categories [27]: those that are intentionally produced and used at an industrial level, and those accidentally formed in processes generally related to human activities, but also related to unpredictable accidental events, such as volcanic eruptions or forest fires. From this point of view, Polychlorinated biphenyls (PCBs) and Polycyclic Aromatic Hydrocarbons (PAHs) are among the most widely studied classes of POPs and have a very interesting feature: PCBs are considered industrially produced chemicals, and PAHs belong exclusively to the accidentally formed POP category. In the first case, once the overall level of contamination has been classified “dangerous to humans”, there is only one possible strategy: to restrict or even ban the industrial use of that compound. In the second case, an effective strategy requires a preliminary assessment of source apportionment (i.e., accidental events and human activities). These pollutants have carcinogenic and mutagenic effects well known for many years. Today they are present in all components of our environment and are contributing significantly to an almost irreversible deterioration of the integrity of our water resources.

## 2.1. Polychlorinated biphenyls (PCBs)

PCBs are a mixture of 209 congeners, according to the number and position of chlorine atoms in the biphenyl skeleton (from monochloro- to deca chloro-biphenyl). They are synthetic products and have been introduced in numerous industrial products since 1930. The estimated quantity produced to date is around 2 million tons of which only 17% has been already eliminated [28]. The main events of the PCB story are the following: 1881: chemical synthesis; 1930: start of industrial production; 1966: first report on the presence of PCBs in the environment; 1970: start of toxicological research and environmental monitoring; 1980: approval of the first laws for the limitation and control in the use of PCBs [29–31]. Because of their high chemical and thermal stability, low flammability and high dielectric constant PCBs were massively used in electrical, hydraulic and heat transfer equipment, and also in pigments, dyes, rubber products, and a lot of other industrial applications [32]. Since the correct application of the guidelines for the use and maintenance of electrical transformers/capacitors containing PCBs is still far from being respected, and the improper discharge of PCB waste is not completely eliminated, there are still many potential sources of emission of PCBs all over the world. Furthermore, the presence of accidentally formed PCBs has been hypothesized in the emissions of industrial and municipal waste incinerators [33,34], and probably also in forest fires [35]. Much more exhaustive information on PCBs can be found at the web site of US-EPA [32].

**Table 1** shows the levels of total PCB concentration in the atmosphere and in the hydrosphere worldwide as a consequence of the ubiquitous characteristics of these contaminants. The references reported are just a few examples of the hundreds of articles published in the last twenty years.

As already mentioned above, PCBs are a class consisting of 209 congeners, and unfortunately it is not always easy to compare literature

data because of the differences in the number of congeners that are included in the total concentration. This is because there are no internationally accepted guidelines. Very often, the total concentration is obtained by adding the concentrations of the following seven congeners which are considered the most abundant in the commercial formulations of PCBs: PCB28 (2,2,4'-trichlorobiphenyl), PCB52 (2,2',5,5'-tetrachlorobiphenyl), PCB101 (2,2',4,5,5'-pentachlorobiphenyl), PCB118 (2,3',4,4',5-pentachlorobiphenyl), PCB153 (2,2',4,4',5,5'-hexachlorobiphenyl), PCB138 (2,2',3,4,4',5'-hexachlorobiphenyl), PCB180 (2,2',3,4,4',5,5'-heptachlorobiphenyl). In a number of publications, especially those related to the effects on human health, the concentration of non-ortho and mono-ortho substituted PCBs is also reported as a consequence of their high toxicity attributed to a quasi-planar configuration which makes them similar to dioxins, and for this they are also called dioxin-like PCBs [60–62].

The concentration levels of PCBs range from fractions of ng/m<sup>3</sup> for the atmosphere and fractions of ng/L for the hydrosphere in remote areas, to values on average 100–1000 times higher in highly populated areas, such as urban areas or the Mediterranean Sea. This is a clear experimental evidence of the direct relationship between the presence of these contaminants and human activities.

## 2.2. Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs belong to the category of POPs accidentally formed during pyrolytic processes, i.e. incomplete combustion of organic material, or originating from petrogenic sources. They enter the environment from numerous sources, mainly through atmospheric deposition, oil spills and industrial wastes. Incomplete combustion of organic material can result from both accidental events such as volcanic eruptions and forest fires [35,63,64], as well as from human activities such as combustion of coal, gasoline and other fossil fuels [65–68], and a variety of combustion processes including industrial and municipal waste incineration plants [69–71].

PAHs never occur individually but in mixtures and, in the absence of internationally accepted guidelines, also for this class of pollutants it is not always easy to compare the literature data because of the differences in the number of compounds included in the total concentration. The United States Environmental Protection Agency (U.S. EPA) selected the following sixteen compounds [72] which are very often reported in environmental monitoring studies: NAP Naphthalene, ACY Acenaphthylene, ACE Acenaphthene, FLU Fluorene, PHE Phenanthrene, ANT Anthracene, FLA Fluoranthene, PYR Pyrene, BaA Benz[a]Anthracene, CRY Chrysene, BbF Benzo[b]Fluoranthene, BkF Benzo[k]fluoranthene, BaP Benzo[a]pyrene, DBA Dibenz[a,h]Anthracene, IPY Indeno[1,2,3-cd]Pyrene, BPE Benzo[ghi]Perylene.

Benzo[a]pyrene, Dibenz[a,h]Anthracene, and a sub-set of six compounds, namely Naphthalene, Benz[a]Anthracene, Chrysene, Benzo[b]Fluoranthene, Benzo[k]fluoranthene, and Benzo[ghi]Perylene, have been classified according to the IARC (International Agency for Research on Cancer) in group 1 (carcinogenic to Humans), group 2A

**Table 1**

The presence of PCBs in the environment.

Environmental component	Location	Concentration range	Ref.
Atmosphere	Antarctica	0.001–0.05 ng/m <sup>3</sup>	[36–40]
	South Pacific/Atlantic	0.01–0.5 ng/m <sup>3</sup>	[39,41,42]
	North Pacific/Atlantic	0.03–0.6 ng/m <sup>3</sup>	[41–43]
	Arctic	0.003–0.6 ng/m <sup>3</sup>	[39,44–46]
	Urban areas	0.1–3 ng/m <sup>3</sup>	[47–49]
Hydrosphere	Antarctica	0.001–0.06 ng/L	[40,42,50,51]
	South Pacific/Atlantic	0.05–0.2 ng/L	[42]
	North Pacific/Atlantic	0.03–0.3 ng/L	[42,43]
	Arctic	0.005–0.2 ng/L	[52]
	Mediterranean Sea	0.1–3 ng/L	[49,53,54]

**Table 2**

The presence of PAHs in the environment.

Environmental component	Location	Concentration range	Ref.
Hydrosphere	Antarctica	0.03–0.3 ng/m <sup>3</sup>	[40,50]
	South Pacific/Atlantic	0.05–1 ng/m <sup>3</sup>	[55,56]
	North Pacific/Atlantic	0.5–5 ng/m <sup>3</sup>	[55]
	Arctic	0.05–1 ng/m <sup>3</sup>	[46]
	Urban areas	1–100 ng/m <sup>3</sup>	[45,57]
	Antarctica	0.05–4 ng/L	[40,51,58]
	South Pacific/Atlantic	0.5–20 ng/L	[55,58]
	North Pacific/Atlantic	0.3–2 ng/L	[43,55]
Atmosphere	Arctic	0.1–0.5 ng/L	[43]
	Mediterranean Sea	1–100 ng/L	[53,54,59]

(probably carcinogenic to human), and group 2B (possibly carcinogenic to Humans), respectively [72]. Benzo[a]pyrene is the only one in group 1, and for this reason its concentration is separately reported and used as a marker of occurrence of carcinogenic PAHs in food [73]. In 2004, the European Union set an annual target for benzo(a)pyrene (BaP) in ambient air of 1 ng/m<sup>3</sup>.[74].

Table 2 shows the levels of total PAHs concentration in the atmosphere and in the hydrosphere worldwide as a consequence of the ubiquitous characteristics of these contaminants. Similarly to PCBs, the references represent a few examples of the hundreds of articles published in the last twenty years.

The mean concentration level of PAHs is less than 1 ng/m<sup>3</sup> for the atmosphere and about 1 ng/L for the hydrosphere in remote areas, and reach levels about 100 times higher in highly populated areas, such as urban areas or the Mediterranean Sea. Very recently, doubts have been raised about the widespread practice of determining only sixteen PAHs, and a proposal has been made to extend the determination to forty PAHs (including the 16 EPA PAHs), twenty three NSO-heterocyclic compounds and six heterocyclic metabolites, and ten oxy-PAHs and ten nitro-PAHs, all included in the class of Polycyclic Aromatic Compounds (PACs) [75]. In conclusion, even for PAHs there is no doubt about the direct relationship between the presence of these contaminants in the environment and human activities.

### 3. Effects of PCBs and PAHs on human health

The environmental hazard and the toxic effects of PAHs and PCBs on biota are now documented by several hundreds of scientific publications, and the awareness and the concern that derives from the effects on human health are now in the public domain. A comprehensive analysis of the literature on this aspect is not included in the aims of this work, and therefore only a few reviews and a few examples are cited with the aim of highlighting the progress of knowledge over the last 40–50 years on these carcinogenic/teratogenic pollutants. A very important aspect common to both classes of pollutants, concerns the ability of these compounds to bio-accumulate along the food chain as a result of their lipophilicity which leads them to concentrate in fatty and lipid-rich tissues. In fact, from levels of ng/L or fractions of ng/L in sea water of areas with a high anthropogenic impact, such as the Mediterranean Sea (Tables 1 and 2), we can find levels as high as µg/kg or hundreds of µg/kg in the tissues of both marine and terrestrial organisms, including humans [76–78].

#### 3.1. Polychlorinated biphenyls (PCBs)

The very first article of the toxic effects of PCBs was published in 1937. This study investigated on the toxic effects of chlorinated compounds, included PCBs “....certainly capable of doing harm in very low concentrations and are probably the most dangerous of the chlorinated hydrocarbons studied” concluded the authors [79]. A lot of reviews and studies followed this first report. As examples, clear damage to liver and

kidney was shown after acute exposure of rats to 1–10 g of Aroclor 1242 per kg of body weight, whereas in the chronic exposure study, 184 rats were exposed to about 5 mg/kg body weight for approximately 21 months, and at the end of experiment 146 of them showed the presence of neoplastic nodules within the liver [80]. Subsequently, it was clarified that the toxic effect of PCBs was congener-dependent [81]. In fact, in a more recent study the greater toxicity of non-ortho- and mono-ortho-substituted congeners was confirmed [82]. A review of animal and human data on PCB exposure cited in the U.S. National Library of Medicine from 2000 to 2010 was also recently published [83]. By way of example, the following data concerning the human species are mentioned: (i) epidemiologic studies of human exposures to PCBs have shown that the incidence of prostate cancer in the high exposure group is about twice the lowest exposure group; (ii) a significant effect on sexual maturation was observed when serum levels of PCB 153 and dioxin-like chemicals doubled; (iii) enamel defects in deciduous teeth significantly high in higher exposed children; (iv) anti glutamic acid decarboxylase (Anti-GAD) was 4 times higher in exposed subjects than that of controls.

Among the numerous papers on PCB exposure, by way of example, the following are cited: (i) association between employment at capacitor manufacturing plants with direct exposure to PCBs and malignant melanoma and cancer of the brain [84]; (ii) excess of mortality from cancers of prostate, stomach and biliary tract, and breast of workers employed at capacitor-manufacturing facilities and exposed to PCBs [85]; (iii) final confirmation of the prostate cancer mortality as a consequence of prolonged exposure to PCBs on an enlarged cohort of 14.458 workers [86].

#### 3.2. Polycyclic Aromatic Hydrocarbons (PAHs)

Evidence for the relationship between the carcinogenic power of PAHs and their link to deoxyribonucleic acid of mouse skin was published more than 50 years ago [87]. Since then, a huge number of articles have been published, confirming reproductive toxicity, cytotoxicity, mutagenicity, and carcinogenicity of PAHs [88]. In 2002, the European Commission's Scientific Committee on Food (SCF) published a report on 33 PAHs potentially occurring in food. The list of major concern for consumers' health included the eight more toxic PAHs selected from the US EPA, and eight additional PAHs [89]. Studies on the effects of exposure to mixtures of PAHs in the workplace have clearly highlighted an increased risk of skin, lung, bladder, and gastrointestinal cancers. Furthermore, the relationship between a long-term exposure to low levels of some PAHs and cancer development has been clearly demonstrated in laboratory animals [90–93]. A review has very recently been published on the effects caused by a chronic exposure to mixtures of PAHs in air. Respiratory effects, ischemic heart disease, chronic dermatitis, depressed immune system have been observed, along with skin and lung cancer. The review conclude that BaP among other PAHs is able to form DNA adducts which are likely the key to their mutagenic and carcinogenic potential to humans [94,95].

#### 4. Source identification and apportionment of PCBs and PAHs

Source apportionment allows the evaluation of the net contribution of human activities to the global environmental pollution. Similarly to the other environmental aspects discussed in this concise review, over the past 30–40 years a very large number of articles have been published concerning source apportionment for PCBs and PAHs. Numerous and complementary approaches have been described and applied to geographical areas with both high and low human impact. As an example, few reviews and few specific studies are reported.

##### 4.1. Polychlorinated biphenyls (PCBs)

The environmental relevance of PCBs is mainly due to their massive industrial use for about 40 years without any concern for their dramatic toxicity and without any precaution to contain their widespread diffusion in the environment. This period dates from 1930, the beginning of industrial production, to 1970, the beginning of the awareness of the serious toxic effects on humans and the approval of the first laws on the restriction of the use of PCBs exclusively in closed systems. According to the EMEP/CORINAIR Guidebook of the European Environmental Agency (EEA), the main sources of PCBs emission into the environment derive from industries producing PCBs, use of equipment and products containing PCBs, emission of sites polluted by PCBs, and combustion processes [96]. Very recently, an exhaustive review of PCBs in US schools has been published. The concern referred to the period of extensive school construction in the United States, as well as in Europe, (between 1950 and 1980) which coincided with the time of the largest use of PCBs as plasticizers in building materials. PCBs levels as high as 100–500 g/kg and 0.1–10 µg/m<sup>3</sup> were found in building materials (caulks and sealants) and indoor air samples, respectively [97]. A further evidence of the widespread presence of PCBs in the environment has been obtained from the high levels of PCBs found in foods (eggs, meat or milk) derived from free range chickens, sheep and beef, in the absence of any known source of contamination [98]. Finally, a very informative Focus Issue of Environmental Science & Technology on PCBs was published in 2010 [99].

Although PCB emission from waste incinerators, forest fires and other accidental sources has been reported (see Section 2.1), a definitive assessment of the relative contribution of the two possible sources (industrial production and accidental formation) has not yet been carried out. An attempt to do so is reported in the section *The message from the South Pole on PCBs and PAHs*, where paleo-environmental studies have been applied for the assessment of source apportionment.

##### 4.2. Polycyclic Aromatic Hydrocarbons (PAHs)

Two main processes are generally considered as PAHs sources: pyrolysis, i.e. incomplete combustion of organic material, and petrogenesis, i.e. decomposition of organic matter by processes occurring in sediments at elevated temperatures and pressures. Pyrogenic sources are characterized by a prevalence of high molecular weight compounds with four to six rings, whereas petrogenic sources by a prevalence of low molecular weight compounds with two or three fused benzene rings. Specific PAH compounds and isomer ratios, such as the ratios of phenanthrene/anthracene and fluoranthene/pyrene, along with matrix factorization and principle components analysis are commonly used to distinguish between petrogenic and pyrogenic sources. Very recently, a review article compared these approaches, and concluded that none of them alone is able to give reliable answers. Thus, in order to minimize the weaknesses inherent in each method, multiple identification techniques should be used to get correct PAH source identification and apportionment [100]. For example, a study was carried out on the coastal atmosphere of Chicago and Lake Michigan. A modified factor analysis-multiple regression model was applied, which showed that combustion of coal, gasoline and other fossil fuels accounted for about

100% the ΣPAH concentration in the atmosphere [101]. Another study that showed the great prevalence of the anthropic contribution concerned the urban environment of Delhi. In this case the Principal Component Analysis (PCA) was applied. The conclusion was that diesel and gasoline driven vehicles were the principal sources of PAH in all seasons, although the combustion of coal and wood contributed significantly to the levels of PAH in winter [102]. A study on the presence of PAHs in riverbank soils of the Mosel and Saar Rivers in Germany was carried out applying several approaches, i.e. the distribution patterns of 45 PAHs, specific PAH ratios, distribution patterns of n-alkanes and principal component analysis (PCA). In this case, the petrogenic input by coal particles was found to be dominated, and was also confirmed that several identification methods must be applied in order to accurately determine the origin of PAHs [103].

#### 5. The message from the South Pole on PCBs and PAHs

Antarctica is a continent almost free of local sources of anthropogenic contamination and is an ideal place to perform paleo-environmental studies on global contamination aimed at assessing both the contribution of human activities and the effectiveness of specific regulatory actions implemented to reduce the level of environmental contamination of a given class of pollutants [63,104–107]. According to their physical-chemical characteristics, PAHs and PCBs can be either in the vapour phase or associated to the atmospheric aerosol [108–113]. Moreover, the low temperatures during the whole year allow the wet deposition of chemical compounds and snow stratification to be obtained, whose chemical composition reflects that of the atmosphere at the time of deposition. This is true only if reliable glacio-chemical data have shown that snow deposition was undisturbed [104,105,114–116]. In this regard, degradation processes, which may occur in the atmosphere during long-range transport of POPs, and post-depositional processes, which occur during the morphological change of snowflakes [37,110,117,118], must be carefully evaluated in the interpretation of experimental data. In fact, the first stage of the transformation process of fresh snow into dense glacial ice takes place with a substantial reduction of the surface area. Moreover, temperature gradients due to the high solar irradiation that occur during summer time may affect the snow/firm metamorphism in the top layer, increasing the permeability for several years after deposition [119]. These processes are responsible for the release of semi-volatile compounds, such as PCBs and PAHs [120], back into the gas phase. Hence, individual concentration of PCBs or PAHs may vary unpredictably, causing discrepancy of the distribution pattern of low and high molecular weight compounds in environmental records between developed and remote/pristine areas. The total concentration of a selected number of compounds for each class is less prone to these artefacts and allows a more reliable evaluation of the experimental data.

Paleo-environmental studies by ice cores have been extensively performed, mainly in Greenland [121] and Antarctica [122,123]. The drilling of snow/firm/ice cores up to a depth of 100–200 m is relatively simple, and generally allows information on the last 3–5 centuries. Deeper drilling requires more sophisticated procedures and the use of an anti-icing liquid to keep the drilling hole open [124]. We collected two ice cores at the Talos Dome and GV7 sites during the XX (2004–2005) and XXIX (2013–2014) Italian scientific expeditions in Antarctica, respectively, through which the depth profiles of PAHs and PCBs were obtained [63,64]. Ice core dating was obtained from data previously published for Talos Dome [125] and GV7 [126]. The area of Terra Victoria (Antarctica) which includes Talos Dome and GV7 is mainly influenced from the North-West (Indian Ocean) and the East (Ross Sea and Pacific Ocean) which account for about 85% of the total precipitations, the remaining coming from the Antarctic plateau [127,128]. This fully supports the primary influence of the most intense well-known volcanic events that have occurred in these areas [125,126]. Table 3 shows the details of sampling, sample processing

**Table 3**

Sampling, sample treatment and instrumental sample analysis procedures for the ice core gathered at Talos Dome and GV7, Antarctica.

Talos Dome	GV7
Ice core drilling	Superficial layer: manual excavation of a 7-m deep trench, collecting fourteen samples of about 7 kg, one every 0.5 m. Deeper layer: drilling by an electro-mechanical system (diameter 100 mm), collecting 32 samples, each about 1.5 m long.
Sample treatment	Samples melt in a pre-cleaned stainless steel container in the clean laboratory of the Italian Base at Terra Nova bay (Antarctica). The samples were immediately extracted twice with 20 mL of n-hexane.
Sample storage	The extracts were stored in glass containers at $-20^{\circ}\text{C}$ until their arrival in Italy.
Instrumental sample analysis	Agilent 7890B gas chromatograph, equipped with a PTV injector, an automatic liquid sampler Agilent 7693A, and an Agilent 7010 triple quadrupole mass spectrometer.
Total PCBs ( $\Sigma\text{PCB}_7$ )	PCB28, PCB52, PCB101, PCB118, PCB138, PCB153, PCB180
Total PAHs ( $\Sigma\text{PAH}_{14}$ )	Acenaphthylene (ACY), Acenaphthene (ACE), Fluorene (FLU), Phenanthrene (PHE), Anthracene (ANT), Fluoranthene (FLA), Pyrene (PYR), Benzo [a]Anthracene (BaA), Chrysene (CRY), Benzo[b]Fluoranthene (BbF), Benzo[k]Fluoranthene (BkF), Benzo[a]Pyrene (BaP), Indeno[1,2,3-c,d]Pyrene (IPY), Benzo[g,h,i]Perylene (BPE)

and instrumental sample analysis procedures.

According to the amount of sample analysed for each data point, i.e. about 1.5 m, the uncertainty on the time scale of about 5 years for the deepest samples and about 3 year for the surface layer was estimated. Several maxima were observed for PAHs and PCBs in both ice cores. The maxima were synchronised with well-known explosive volcanic eruptions, and attribution of peak concentrations to specific eruptions was in very good agreement with the concentration of non-sea-salt sulphates which is a well-known marker [63]. Although this result was expected for PAHs, it represents the first clear experimental evidence of the emission of PCBs in the environment during volcanic eruptions. Moreover, Talos Dome temporal profile showed a very intense peak around 1815 which was attributed to the Tambora eruption (Lesser Sunda 1816, explosive index = 6) and supported also by the highest peak concentration of non-sea-salt sulphates [125]. In conclusion, volcanic eruptions were responsible of PCB and PAH peak concentrations in the atmosphere, but the increase resulted very limited in time.

Fig. 2 shows the temporal trend of both classes of pollutants for roughly the last 120 years (1900–2014), as obtained by both Talos Dome and GV7 ice cores. PCBs showed a very high increase in the period 1950–1990 as a signature of the anthropogenic activities due to the massive industrial use of these compounds [129,130].

From 1990 to 2014, a clear trend to the reduction of the level of PCBs was observed which was in very good agreement with the very first experimental evidence of this tendency previously reported for the ice core gathered at Talos Dome [63]. The same decreasing trend was observed also in ice cores collected at the Lomonosovfonna glacier (Svalbard, North Pole) [131], and at the Fiescherhorn glacier (Switzerland) [132]. This result can be considered a direct consequence of the actions taken in many countries to ban the production of PCBs or to restrict their use to closed systems. In particular, the UNECE Protocol on Persistent Organic Pollutants (POPs) required to all parties to eliminate equipment containing more than 50 ppm PCBs by 2015, as reported by the Convention for the Protection of the Marine Environment of the North-East Atlantic (the “OSPAR Convention”) [133].

On the contrary, the level of PAHs showed a clear tendency to increase in the whole observed period of time, with a slope of 0.008 and 0.011 ng/L per year at GV7 and Talos Dome, respectively. The increase is in agreement with data published on ice-cores collected at Mt. Everest [134,135]. Very different trends have been observed in two ice cores collected in September 2003 on Colle Gnifetti, Monte Rosa massif, in the Swiss/Italian Alps [136]. While the level before 1900 was very low (about 2 ng/L) and comparable to those observed in Antarctica in the same period, starting from 1930 a progressive increase was observed in alpine ice cores that was attributed to the rapid industrial development, reaching a maximum of about 30 ng/L in the period 1945–1955.

Moreover, the concentration peaks observed in the Antarctic ice cores in correspondence of specific volcanic eruptions fit very well the

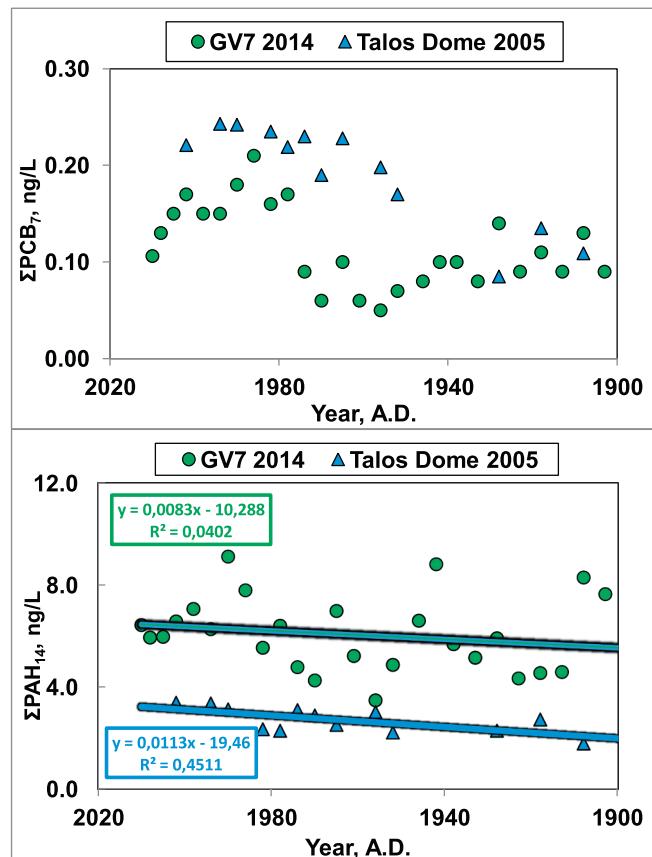


Fig. 2. Temporal trend of PCBs and PAHs for the last 120 years (1900–2014) at Talos Dome and GV7 (Antarctica).

dating of the ice core. The rapid decrease to the background values, observed before and after the maximum, indicates equally clearly a predominant anthropic contribution on the background level due to the long distance transport processes from the more industrialized areas.

In conclusion, the increase in the environmental level of PCBs that was observed for 1930–1980 in the ice cores of the South Pole can be almost totally associated with the production and massive use of these compounds. The slight decrease in PCBs in the last 10–15 years is attributed to the reduction of industrial PCB production and to the limited use only in closed systems. But the global level of PCBs is still too high and the estimated reduction trend is too slow. Therefore, much more international efforts are required to further reduce the spread of PCBs with the ultimate goal of the total ban on their use and production. On the other hand, ice cores evidenced a constant increase of the

**Table 4**

Mean concentration of PCBs and PAHs in human breast milk from various sampling area.

Sampling area	No. of subjects	PCBs ( $\mu\text{g/L}$ )	PAHs ( $\mu\text{g/L}$ )	Ref.
Hong Konk (China)	30	0.2 (Total PCBs)	8.0 ( $\Sigma\text{EPA-PAH}_{16}$ )	[137]
Mersin, (Turkey)	47	0.03 ( $\Sigma\text{PCB}_6$ )	0.35 ( $\Sigma\text{EPA-PAH}_{16}$ )	[138]
Karvina, (Czech Rep.)	191	–	2.9 ( $\Sigma\text{EPA-PAH}_{16}$ )	[139]
Ceske Budejovice, (Czech Rep.)	143	–	2.0 ( $\Sigma\text{EPA-PAH}_{16}$ )	[139]
London and Lancaster, UK	27	0.7 ( $\Sigma\text{PCB}_{15}$ )		[140]

The concentrations are expressed as mg/L of milk with a mean fat content of 4% [136–138]

Total PCBs = all congeners found;  $\Sigma\text{PCB}_6 = \text{PCB}28 + 52 + 101 + 138 + 153 + 180$

$\Sigma\text{PCB}_{15} = \text{PCB}28 + 77 + 99 + 105 + 118 + 138 + 153 + 156 + 167 + 170 + 180 + 183 + 187 + 194 + 203$

environmental level of PAHs associated to an increase of combustion processes involved in many human activities, i.e. combustion of coal, gasoline and other fossil fuels, industrial and municipal waste incineration plants. At present, the effort to adopt international actions to reduce combustion processes has not produced any appreciable results.

## 6. Conclusions

The path to a conscious management of water eco-systems clearly demonstrates that the integrity of the water resources is strongly compromised by the high level of chemical pollution. In fact, the comprehensive analysis of all data available in literature for PCBs and PAHs, which have been taken as representative examples of the POPs, has clearly assessed the negative impact on the level of ecological integrity of water resources, along with a very high risk for human health. In order to make this last statement even more dramatic, Table 4 shows the levels of PCBs and PAHs in breast milk samples from various areas of our planet expressed in  $\mu\text{g/L}$  [137–140], assuming a mean fat content of 4% [141–143].

There is no need to stress the implications of these data: the primary food of our newborns in their first months of life is the main source of exposure to very high levels, i.e.  $\mu\text{g/L}$ , of carcinogenic and teratogenic substances, and studies on source identification and apportionment have unequivocally demonstrated the contribution from human activities on the environmental level of these substances. On the other hand, there is an international sharing of the principle that the first preventive safeguard of human health concerns the integrity of the water resource, and more generally the quality of the environment. Therefore, in a rational system that has adopted this principle, the approval of actions to try to mitigate the negative effects of human activities on the environmental quality should be immediate.

Unfortunately, most of the time, research and management are considered separate activities, implemented in the absence of any level of integration. Adapting management to new information received from research and monitoring should be the right path to follow. From this point of view, the message from the South Pole is crystal clear and the risk to human health is well assessed, but do policy makers do their job well?

To answer this question, let's take into consideration the results of the world's most important meeting on these topics: the meeting of the Triple Conferences of the parties (COPs) to the Basel, Rotterdam and Stockholm conventions. The first extraordinary meeting was held in Bali, Indonesia, from 22 to 24 February 2010. Subsequently, the COPs meeting was held permanently in Geneva, Switzerland, from 28 April to 10 May 2013, from 4 to 15 May 2015, from April 24 to May 5, 2017, and has already been scheduled from April 29 to May 10, 2019. More than 1000 people attended these meetings with a substantial economic and organizational effort. Although it has been nine years since the first meeting and despite all the efforts made so far, The Triple Conference of Parties (Triple COPs) appears to have failed in its objective of “*translating science into action*”. In fact, in the introduction of the 2019 report is written the following: “*At their meetings in 2017, the Triple COPs adopted identical decisions entitled “From science to action”, by which,*

*among other things, they took note of a draft road map, prepared by the Secretariat, for further engaging Parties and other stakeholders in an informed dialogue for enhanced science-based action in the implementation of the conventions.”* [144]. Unfortunately, at the end of the report some of the so-called proposed actions were the following:

- “*The Conference of the Parties*
- *Takes note of the revised draft road map for further engaging Parties and other stakeholders in informed dialogue for enhanced science-based action in the implementation of the conventions;*
- *Encourages Parties and others to initiate action that promotes the implementation of the road map;*
- *Invites Parties and observers to submit to the Secretariat, by 31 August 2020, information on action being undertaken to promote the implementation of the road map;*
- *Requests the Secretariat to continue to cooperate and coordinate with the United Nations Environment Programme and, as appropriate, other relevant organizations, scientific bodies and stakeholders towards strengthening the science-policy interface.”*

In other words: nothing really effective. Many of the decisions up for adoption – which were supported by the vast majority of parties – were blocked by a handful of countries, or in some cases just one. This unfortunately means that at present the integrity of water resources and, more generally, the environment and human health are not of primary importance for our policy makers.

## Acknowledgements

The financial support of the ENEA-National Antarctic Research Program (PNRA) is gratefully acknowledged.

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